Tailoring spin orientation via manipulating electronic structure in two-dimensional magnet CrI_3

Aug. 28, 2019 at IBS

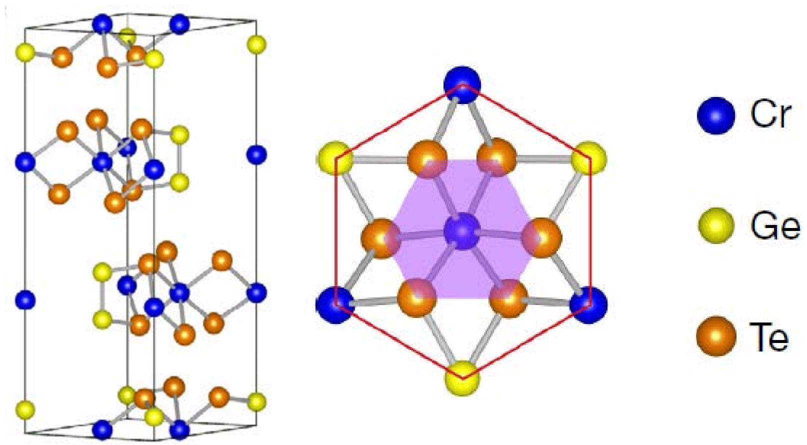
Jeongwoo Kim

Department of Physics, Incheon National University, Incheon 22012, Korea

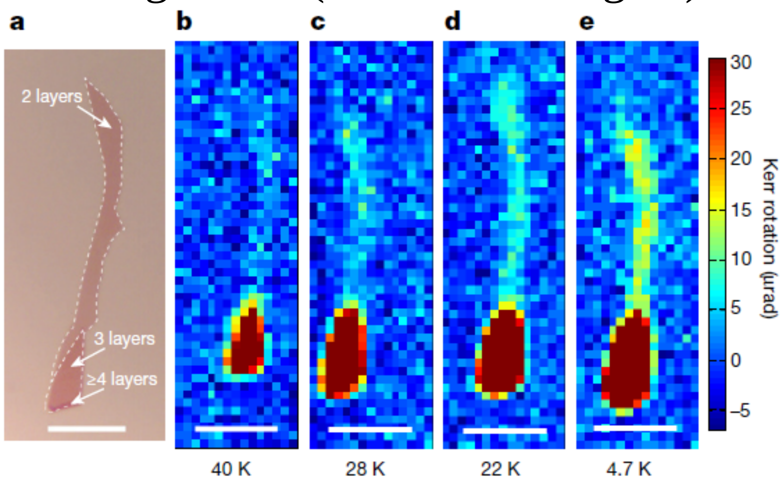
Discovery of two-dimensional magnets

$Cr_2Ge_2Te_6$

Atomic structure



Ferromagnetism (Ker rotation signal)



C. Gong *et al.*, *Nature* 546 265 (2017)

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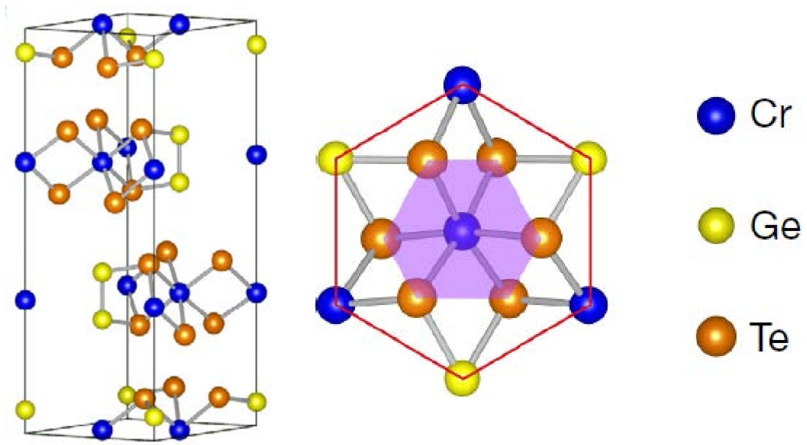

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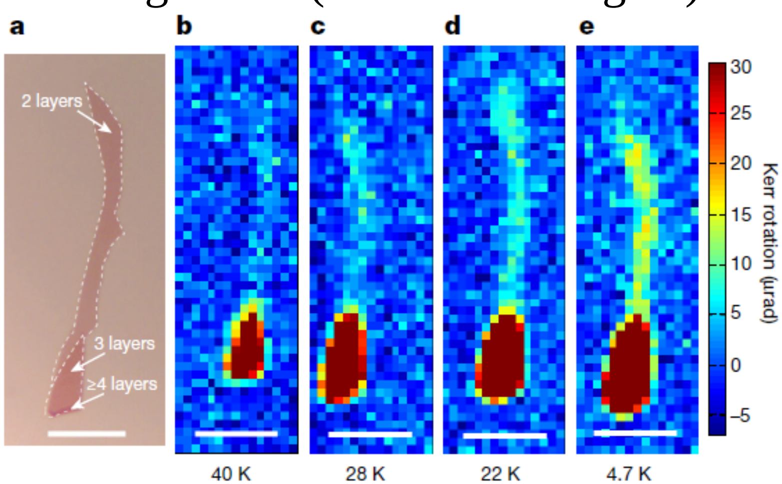

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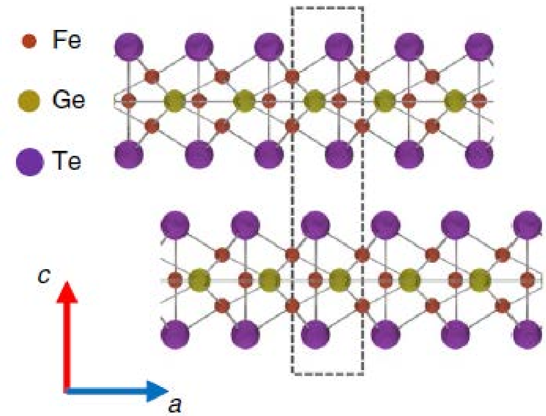
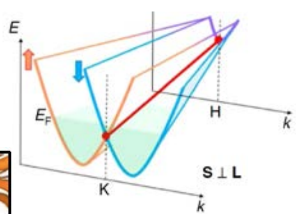
Fe_3GeTe_2

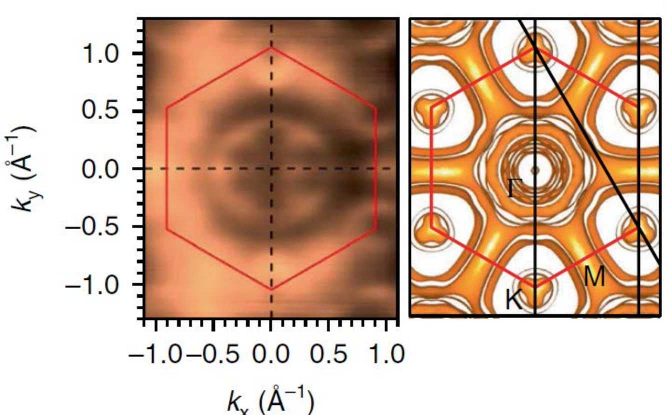
Atomic structure



Z. Fei *et al.*, *Nat. Mater.* 17 778 (2018)

Topological nodal line





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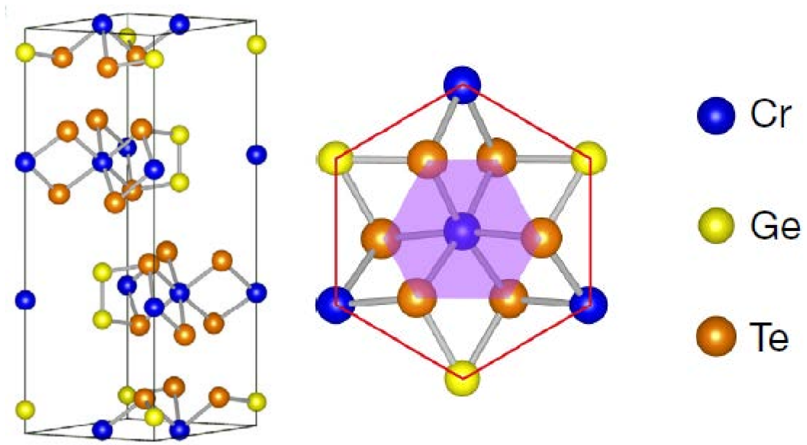

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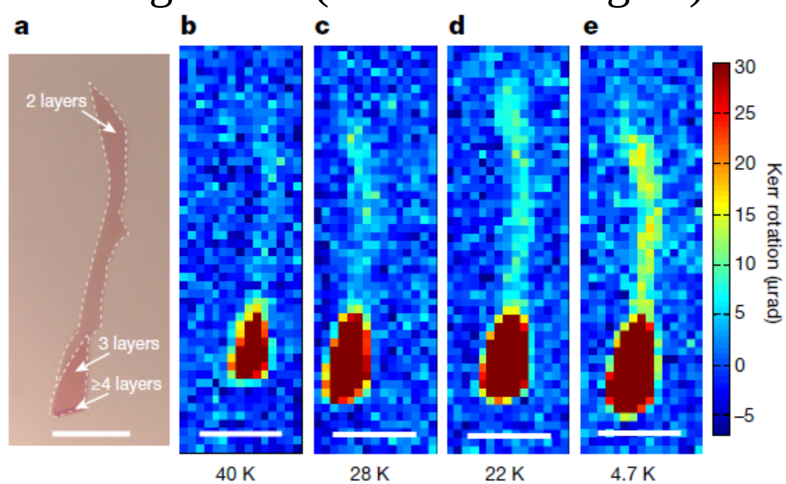

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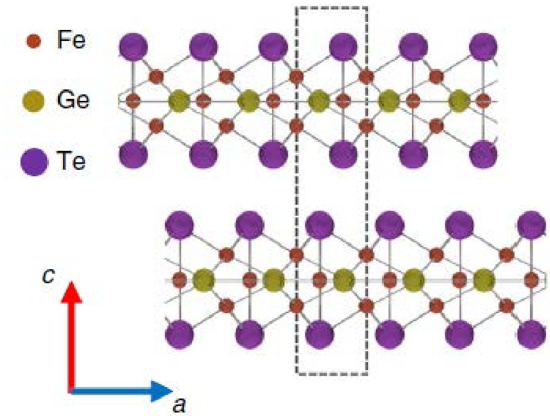

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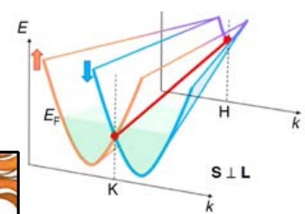

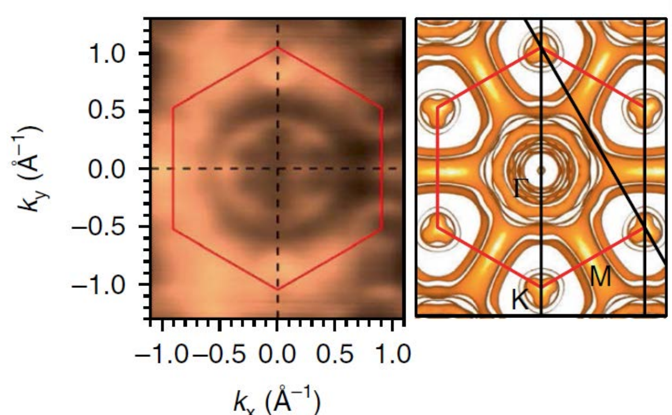

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Discovery of two-dimensional magnets

CrI_3

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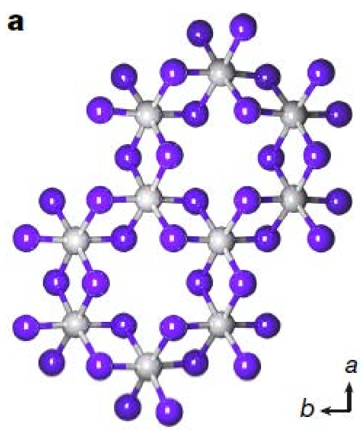
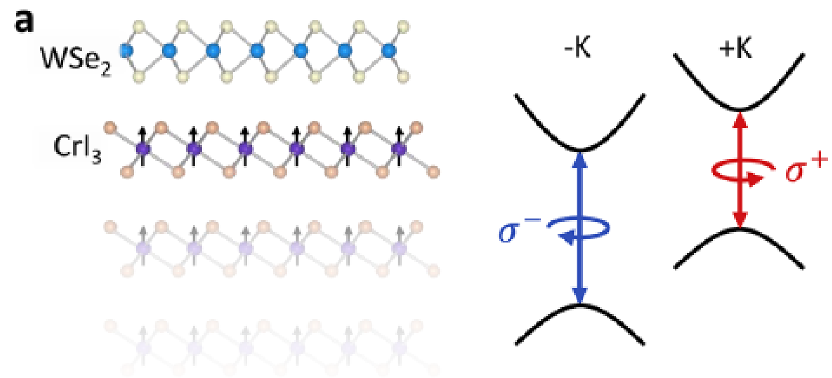
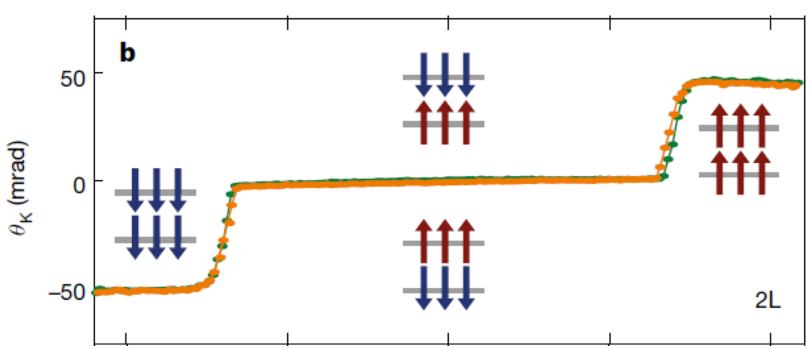


Optoelectronic application (WSe2/CrI3)



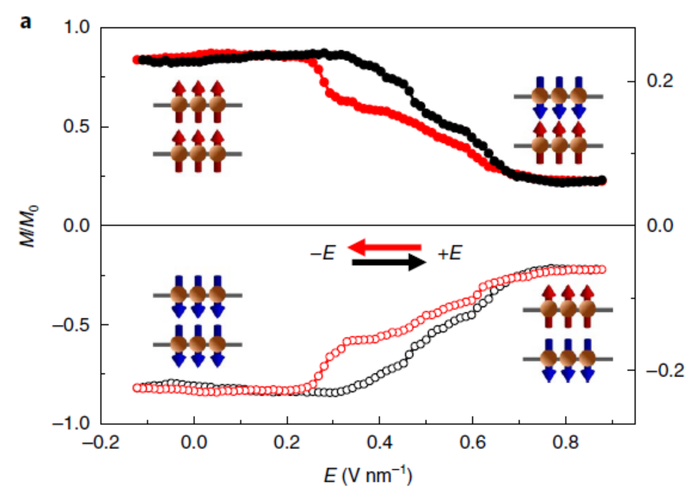
K. L. Seyler *et al.*, *Nano Lett.* 18 3823 (2018)

Antiferromagnetic inter-layer coupling



B. Huang *et al.*, *Nature* 546 270 (2017)

Electrostatic control of the inter-layer coupling

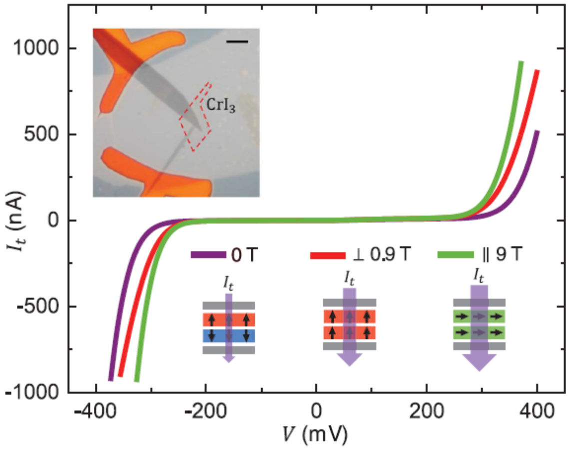


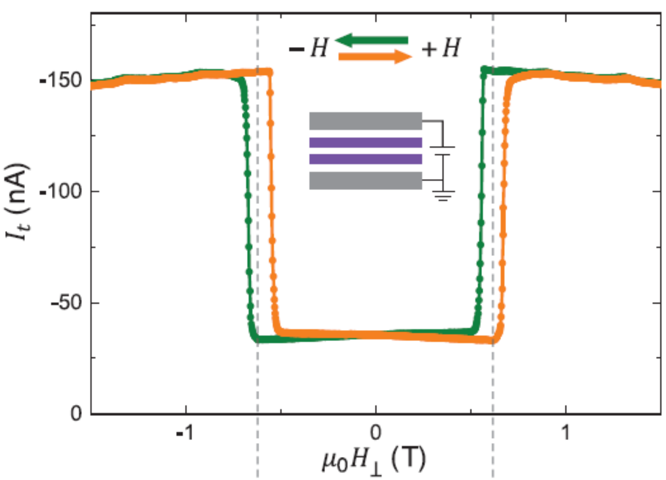
S. Jiang *et al.*, *Nat. Mater.* 17 406 (2018)

Discovery of two-dimensional magnets

CrI_3

Giant tunneling magnetoresistance





T. Song *et al.*, *Science* 360 1214 (2018)

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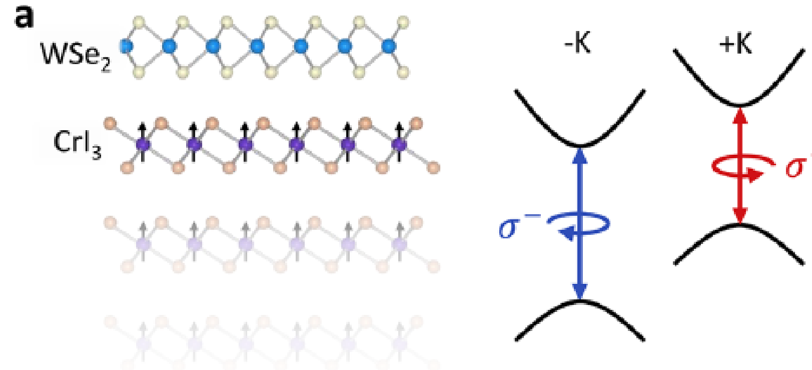

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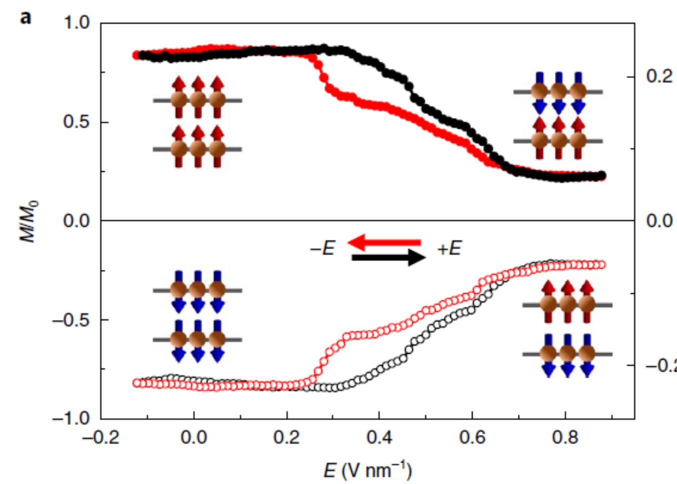

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Magnetic Anisotropy Energy in 2D

The 2D Heisenberg model, in which the spins can point in any direction, does not have a state with long-range magnetic order.

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⇨ **The magnetic anisotropy** in reduced dimensionality is an essential ingredient for the presence of the 2D magnet.

Magnetic Anisotropy Energy in 2D

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⇨ **The magnetic anisotropy** in reduced dimensionality is an essential ingredient for the presence of the 2D magnet.

For physics : an in-depth investigation is prerequisite to the understanding of the underlying physics of the 2D magnetism.

The source of the magnetic anisotropy in CrI_3

i) Ferromagnetic super-exchange (Major)

ii) The single ion anisotropy (Minor)

J L Lado *et al*., *2D Mater.* 4 035002 (2017)

Magnetic Anisotropy Energy in 2D

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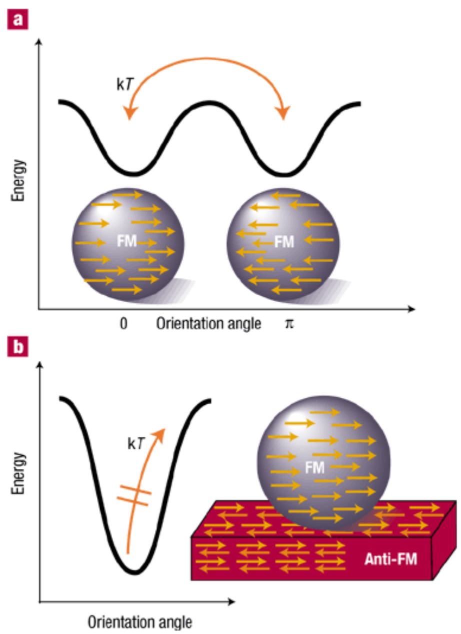
⇨ **The magnetic anisotropy** in reduced dimensionality is an essential ingredient for the presence of the 2D magnet.

For application: the dynamics control and optimization are indispensable



1. MAE$\propto$stability of stored information (Good)

2. MAE$\propto$energy consumption for writing information (Bad)

J Eisenmenger *et al.*, *Nat. Mater.* 2 437 (2003)

My questions are …

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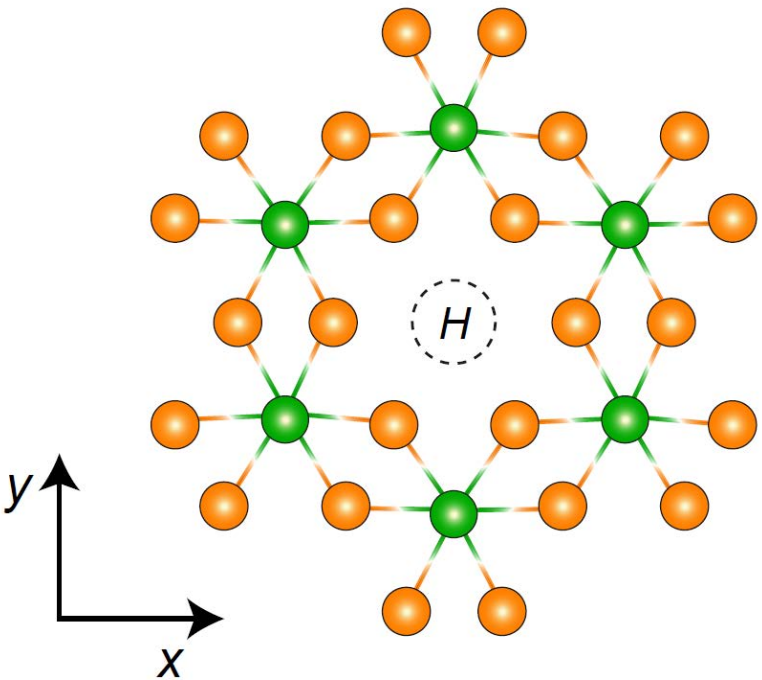
⇨ **Enhanced stability and Low-energy operation**

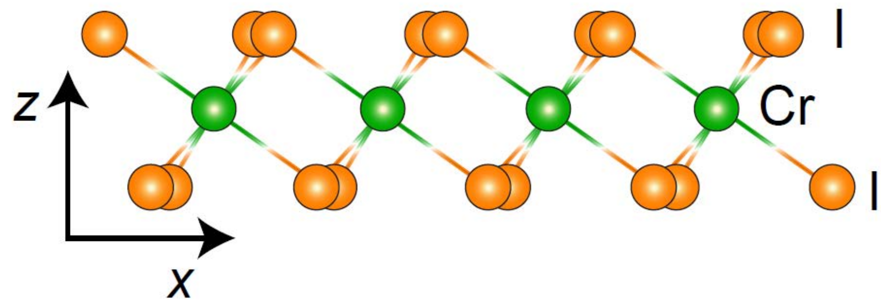
Atomic structure of CrI_3

Top view



H: hollow site

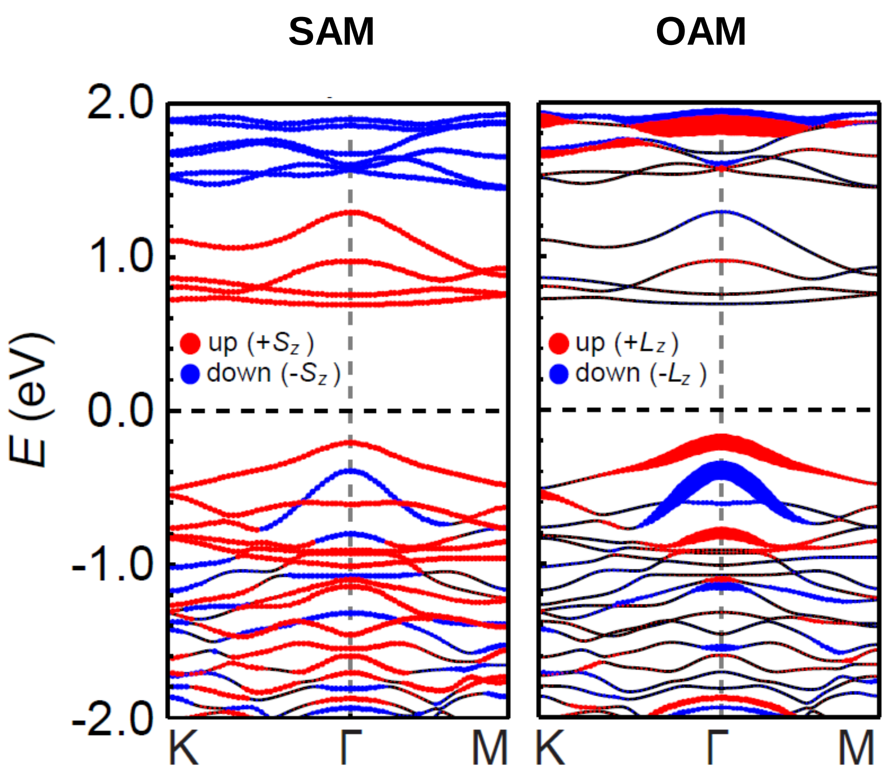
Side view



D_{3d} point group

Electronic structure of CrI_3

Calculated band structure with spin and orbital angular momentum resolution



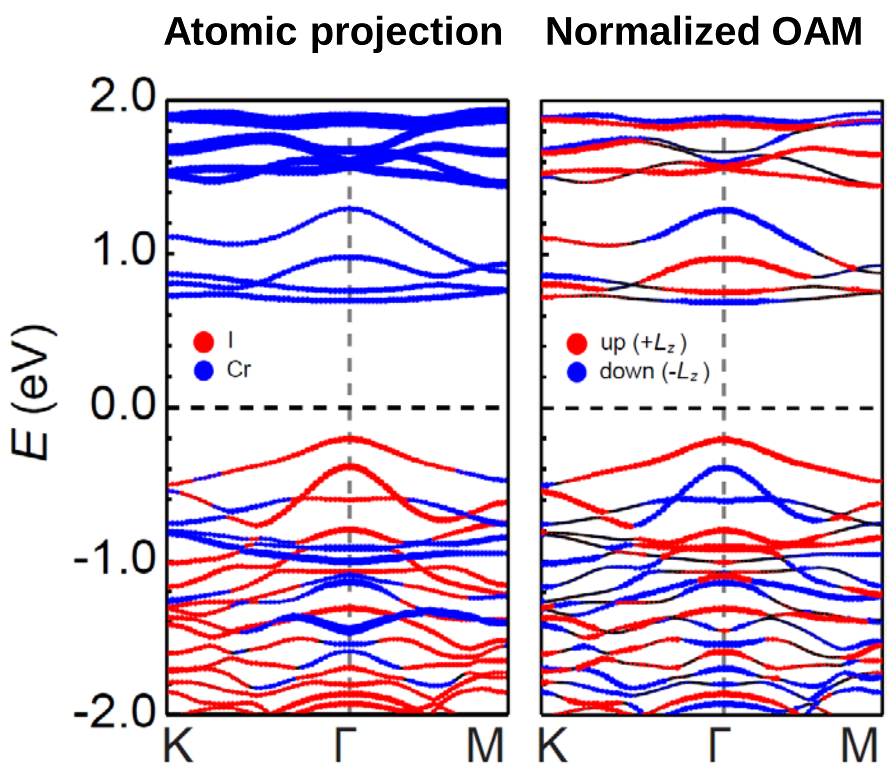
- Cr atom has 3 μ_B spin magnetic moment
- Ferromagnetically ordered with the out-of-plane easy axis
- Spin-polarized band structure (left)
- Substantial orbital angular momentum $\langle L_z \rangle$ of two highest valance bands

two highest valance bands (HVBs)

Two lowest conduction bands (LCBs)

Electronic structure of CrI_3

Calculated band structure with atomic projection and normalized OAM



$L_z/[L_x^2+L_y^2+L_z^2]^{1/2}$

- The orbital angular momentum mainly originates from *p* orbitals in I atoms.
- $\langle L_x\rangle, \langle L_y\rangle \ll \langle L_z\rangle$
- The discrete rotational symmetry around *z* axis implies that L_z is a good quantum number.

Electronic structure of CrI_3

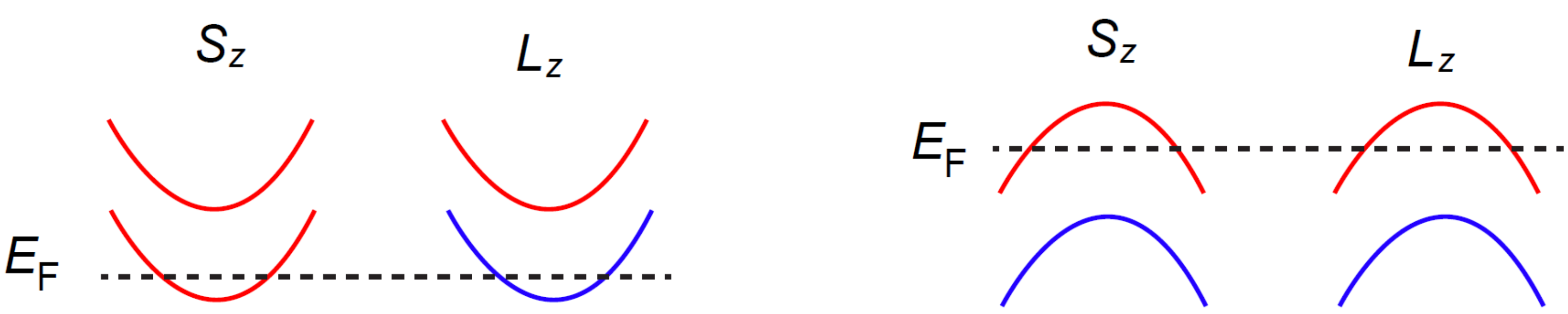
Evaluation of MAE

$$\mathrm{MAE} = \xi^2 \sum_{u,o,\sigma,\sigma'} \sigma\sigma' \frac{|\langle o,\sigma|L_z|u,\sigma'\rangle|^2 - |\langle o,\sigma|L_x|u,\sigma'\rangle|^2}{E_{u,\sigma} - E_{o,\sigma'}}$$

D.-S. Wang *et al.*, *Phys. Rev. B* 47 932 (1993)

ξ : the spin-orbit coupling constant for electrons in Cr
u and o : unoccupied and occupied states
$E_{u/o,\sigma}$: the energy of the states
the spin indices σ and σ'

n doping — *p* doping



- The overlap between the two bands with different $\langle L_z \rangle$ values is not significant.
- L_x operator can mix the two HVBs (LCBs)

$$\Rightarrow \quad |\langle o,\sigma|L_z|u,\sigma'\rangle|^2 - |\langle o,\sigma|L_x|u,\sigma'\rangle|^2 < 0$$

Electronic structure of CrI_3

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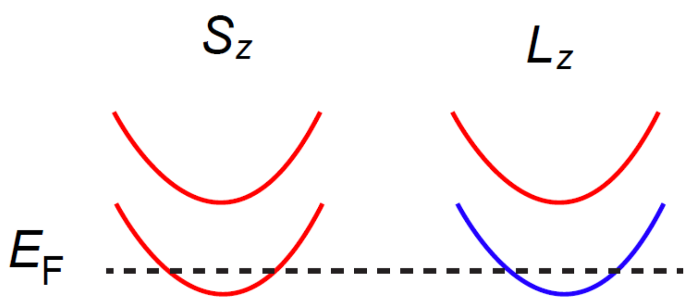
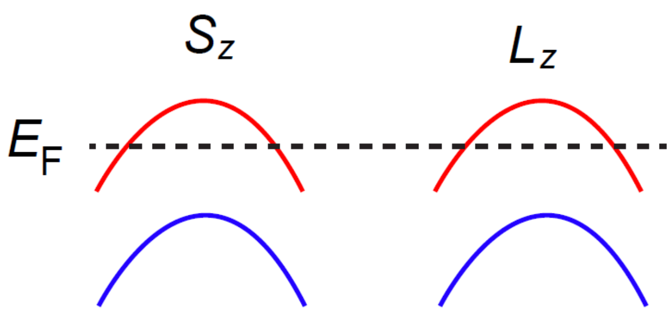


Same spin polarization → negative MAE
(in-plane)

p doping



different spin polarization → positive MAE
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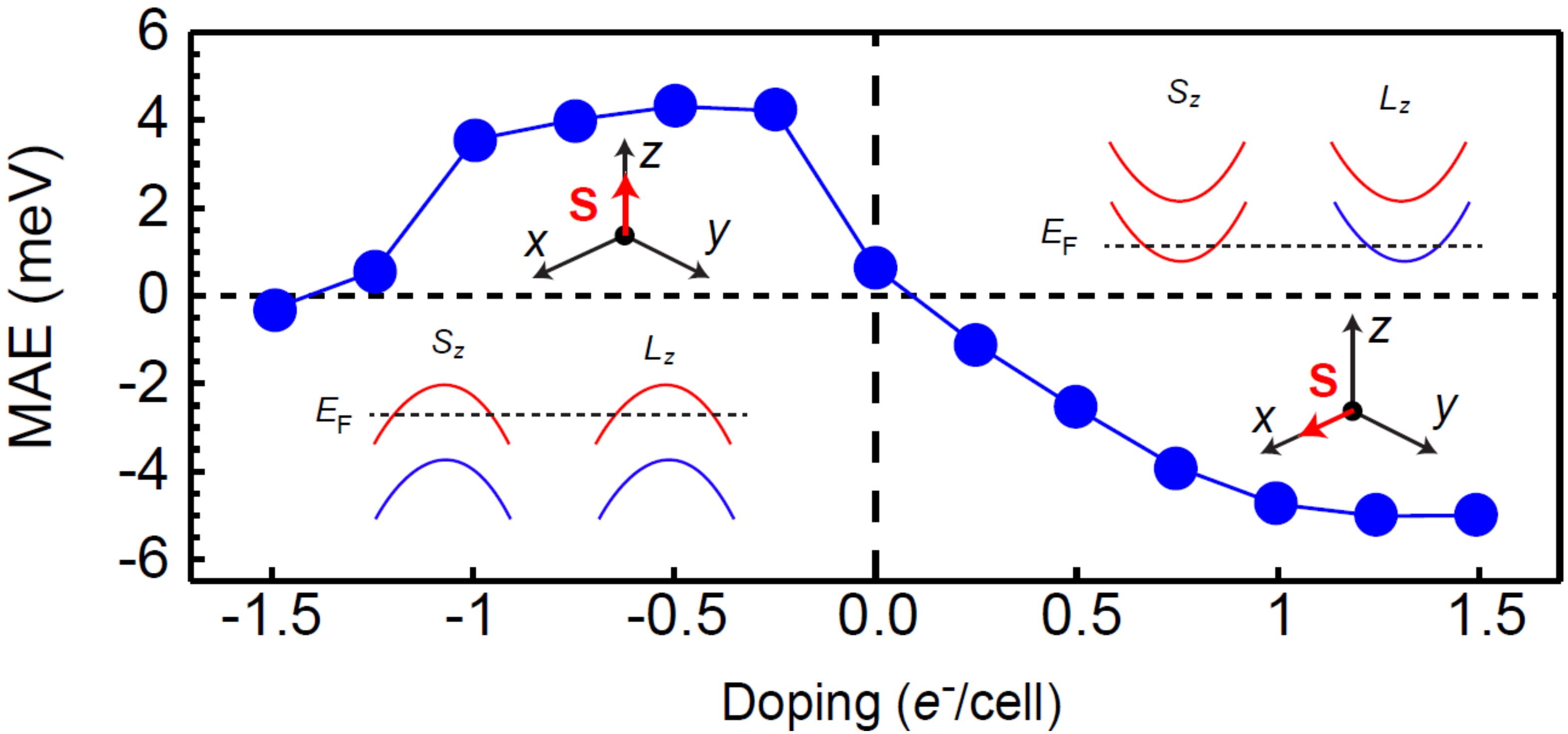
Electronic structure of CrI_3

Calculated MAE with various doping levels

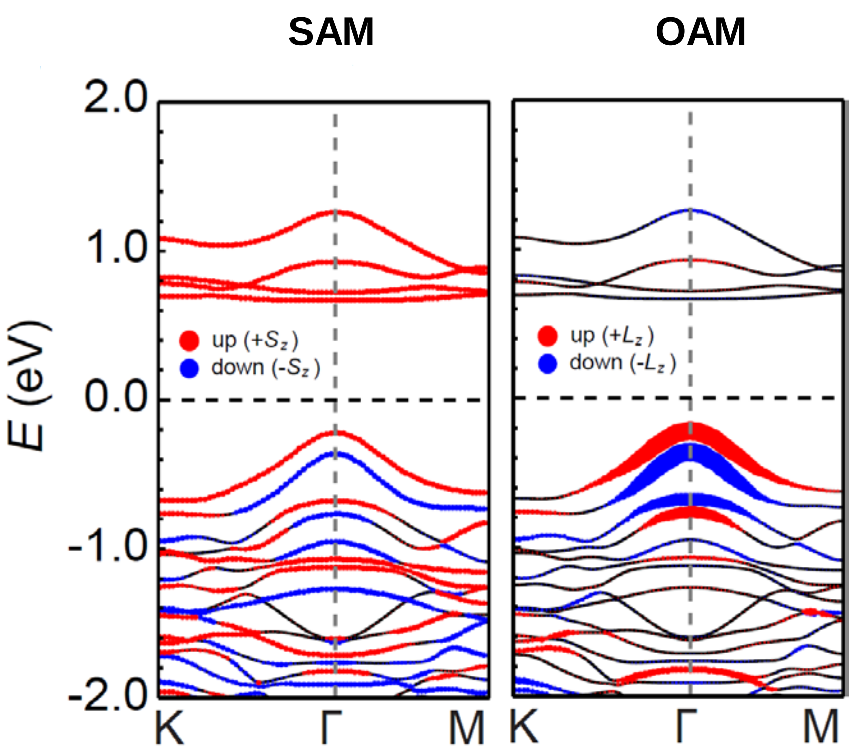


- The MAE increases up to 4.35 meV from 0.66 meV (~ + 560 % change) by reducing electrons.

- It becomes negative and saturated to -5.01 meV (~ -860 % change) by adding electrons.

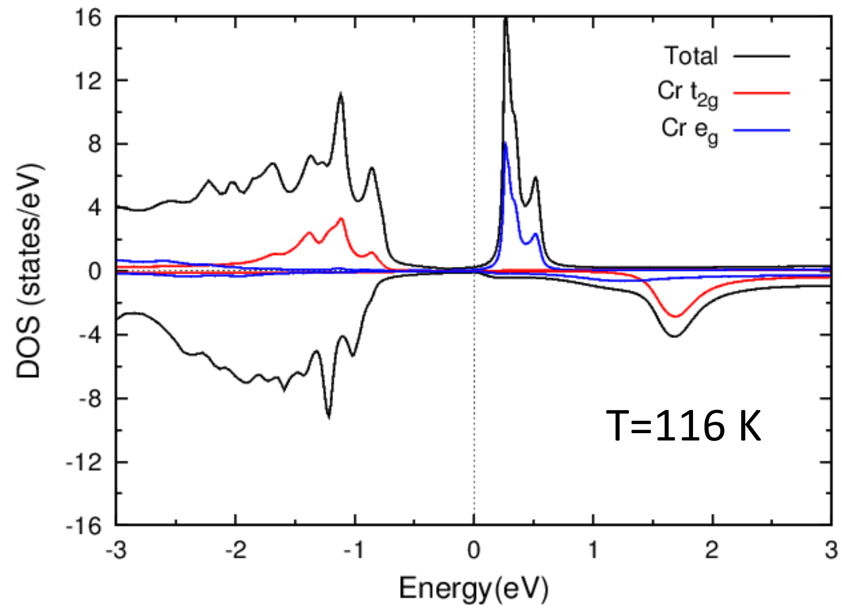
correlation effect

GGA+*U* scheme



The unique angular momentum distributions of the CrI_3 are retained even though the *U* effect (2.65 eV) is considered

DMFT calculations

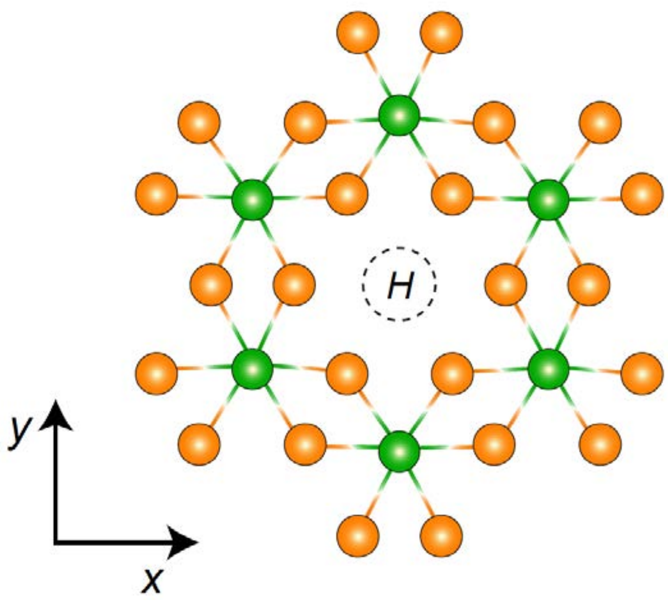


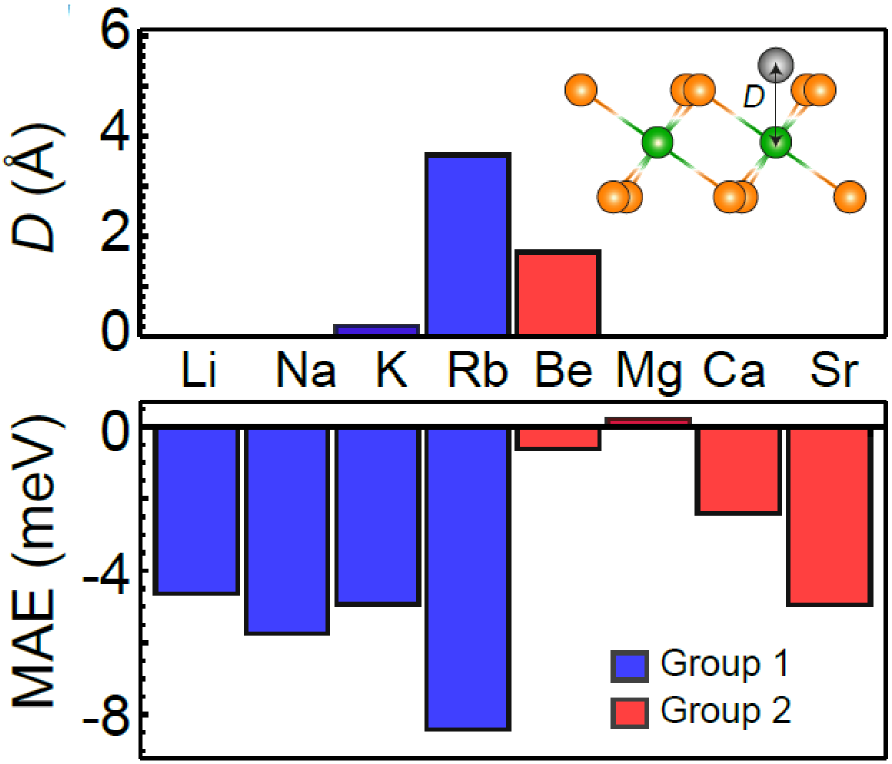
-The hybridization expansion continuous-time quantum Monto Carlo

- Ising-type Coulomb interaction with the on-site Coulomb repulsion U = 10 eV and the Hund's coupling J = 1 eV in Cr 3d orbital

Metal adsorption

Alkali and Alkaline-earth metal adsorption



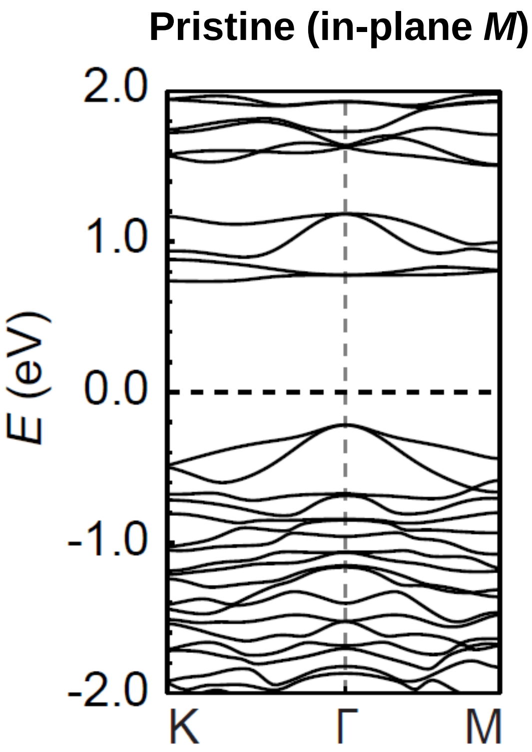


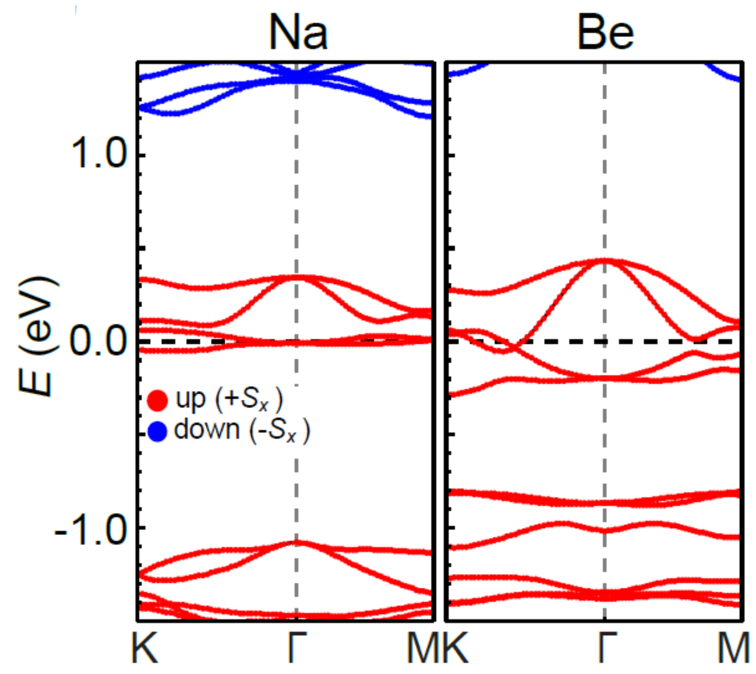
- Alkali metals: yielding sizeable MAEs corresponding to the electron addition case.

- Alkaline-earth metals: deviating from the ideally electron-doped one.

Metal adsorption

Electronic structures



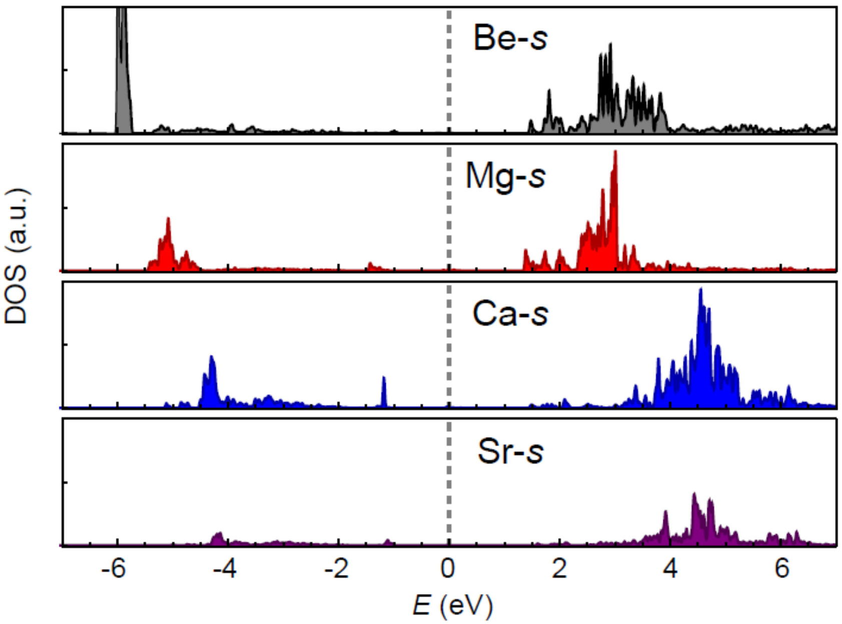


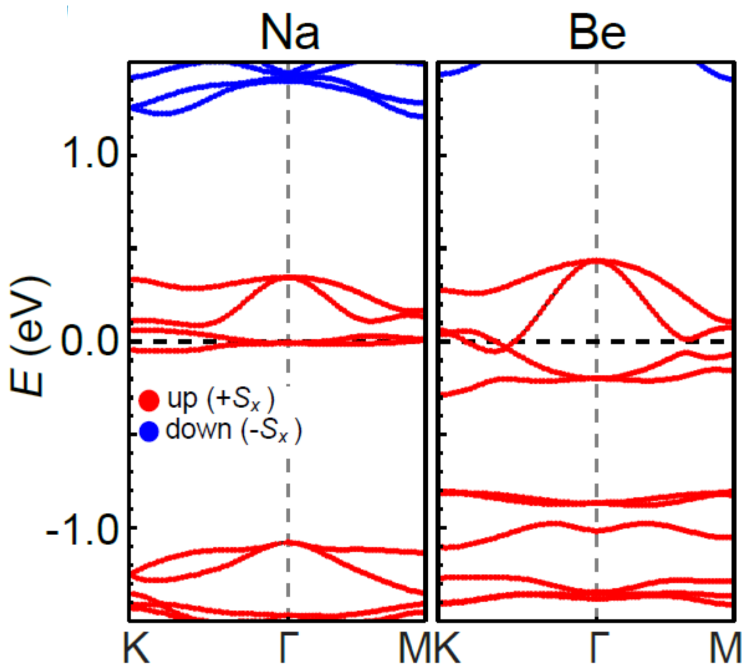
- For the Na adsorption, the band structure remains almost rigid except for the shift of the Fermi level caused by the charge transfer.

- On the other hand, Be atom considerably changes the electronic structure of CrI_3.

Metal adsorption

Electronic structures





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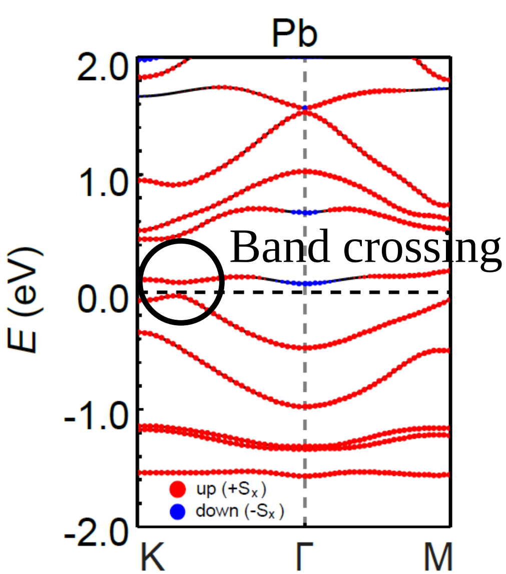
Metal adsorption

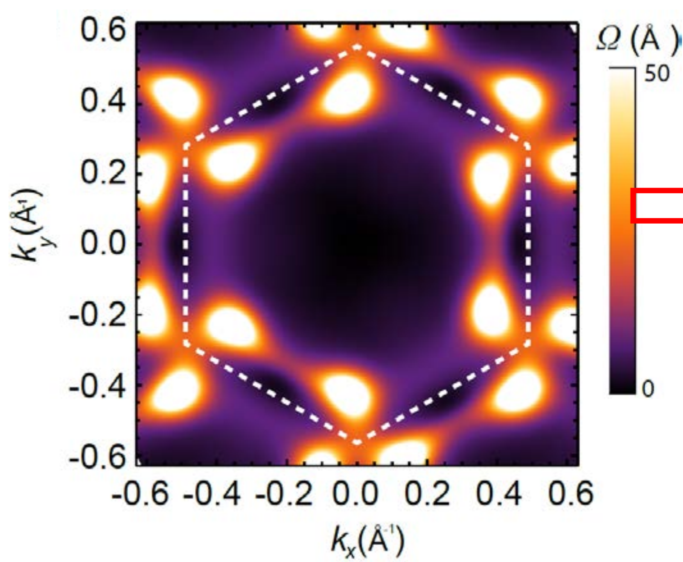
Importance of magnetic anisotropy in band topology

The magnetic anisotropy is very sensitive to external perturbations and thus the ground magnetic anisotropy may not be maintained in 2D magnets, sometimes leading to a misleading result without the consideration of the modified spin axis.

Example: Pb-adsorbed CrI_3



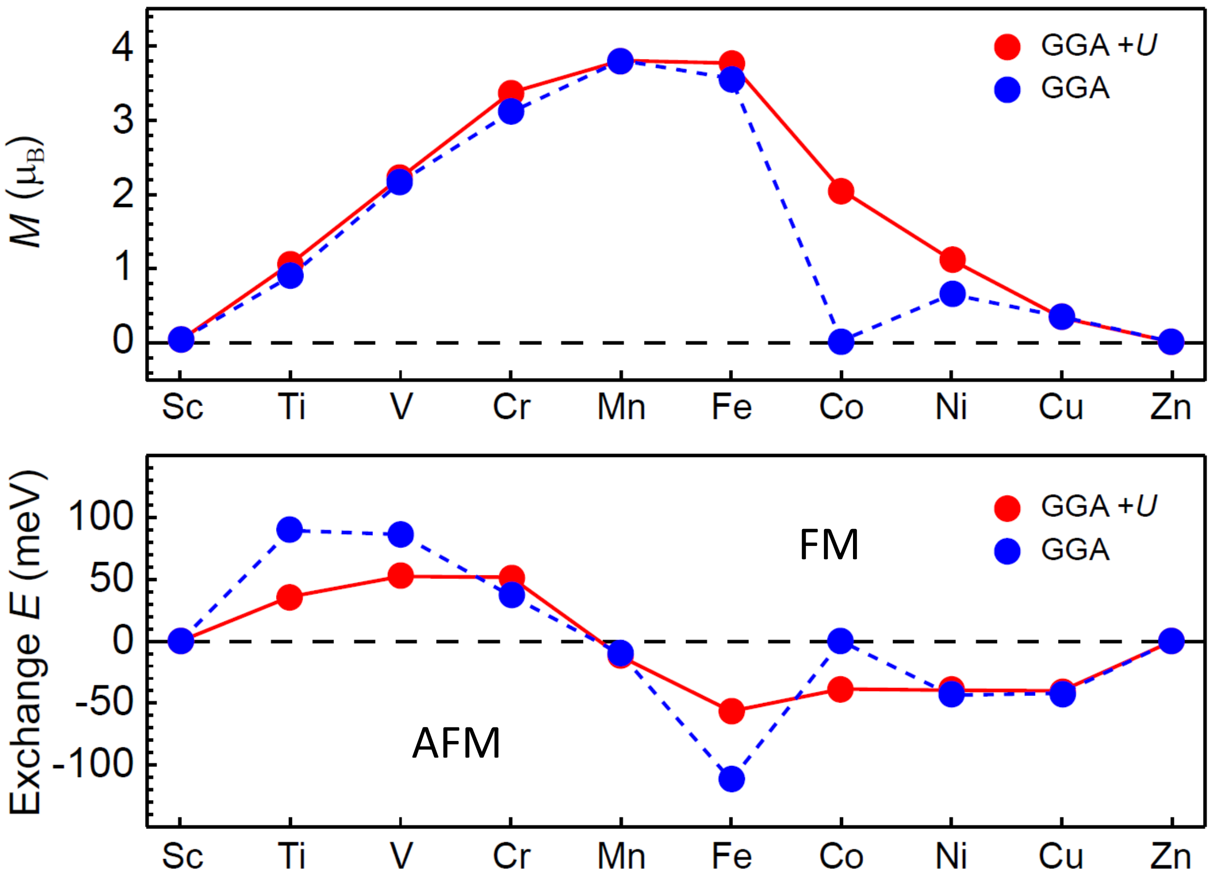
Quantized Berry curvature Flux



- a Chern insulator under the out-of-plane magnetic axis
- indeed a trivial small-gap insulator with the energetically stable in-plane magnetization

Substitutional doping

Local magnetic moment and exchange energy



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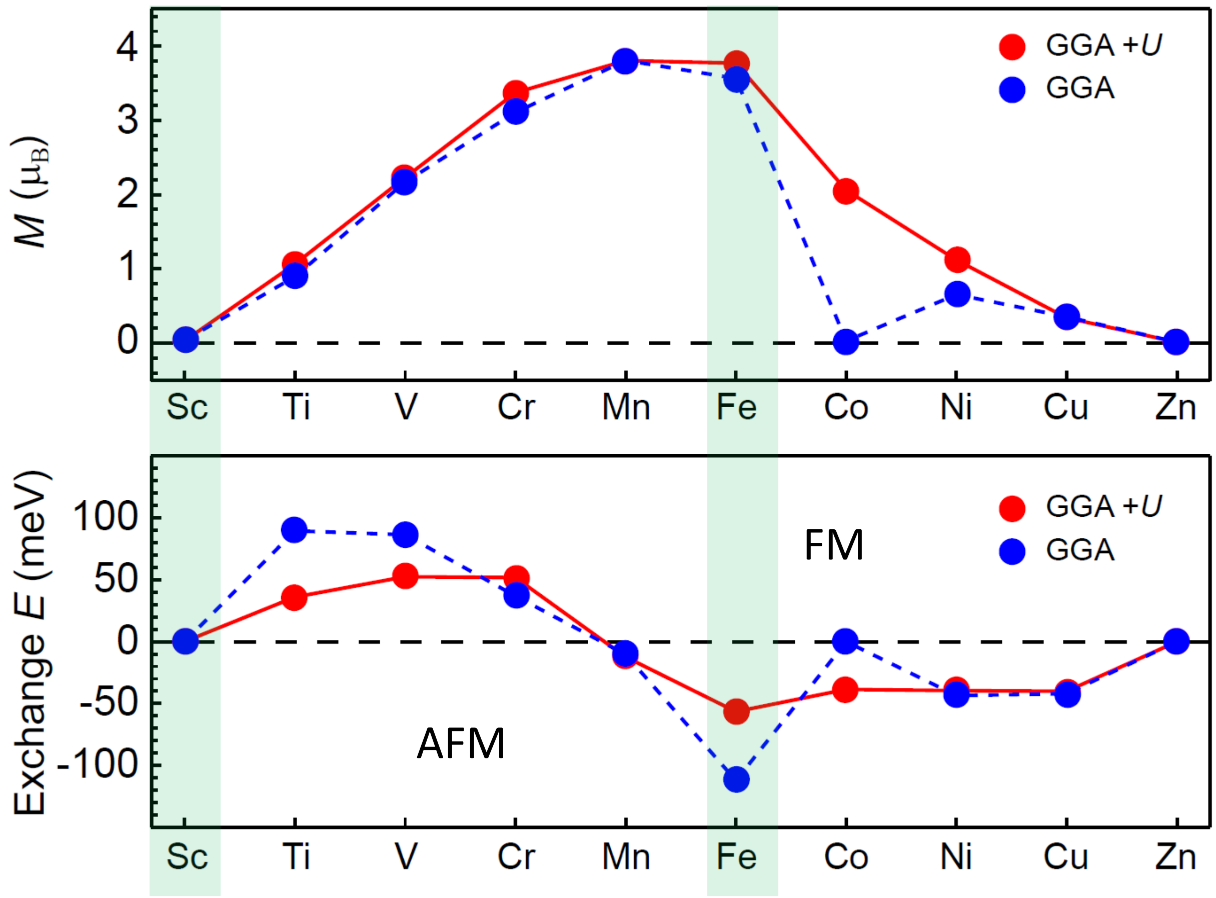
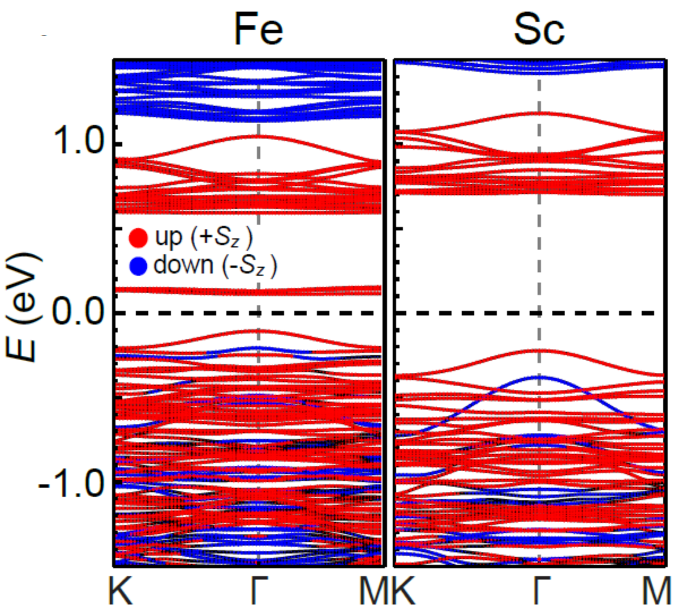


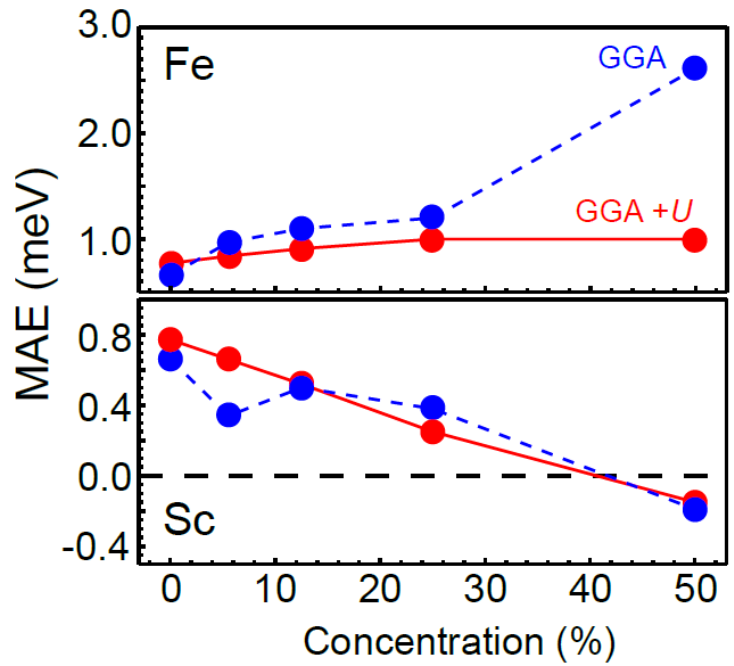
Among 3*d* transition metals, Fe and Sc atoms maintain the insulating nature of CrI_3 when they are substituted for Cr atoms.

Substitutional doping

Band structure



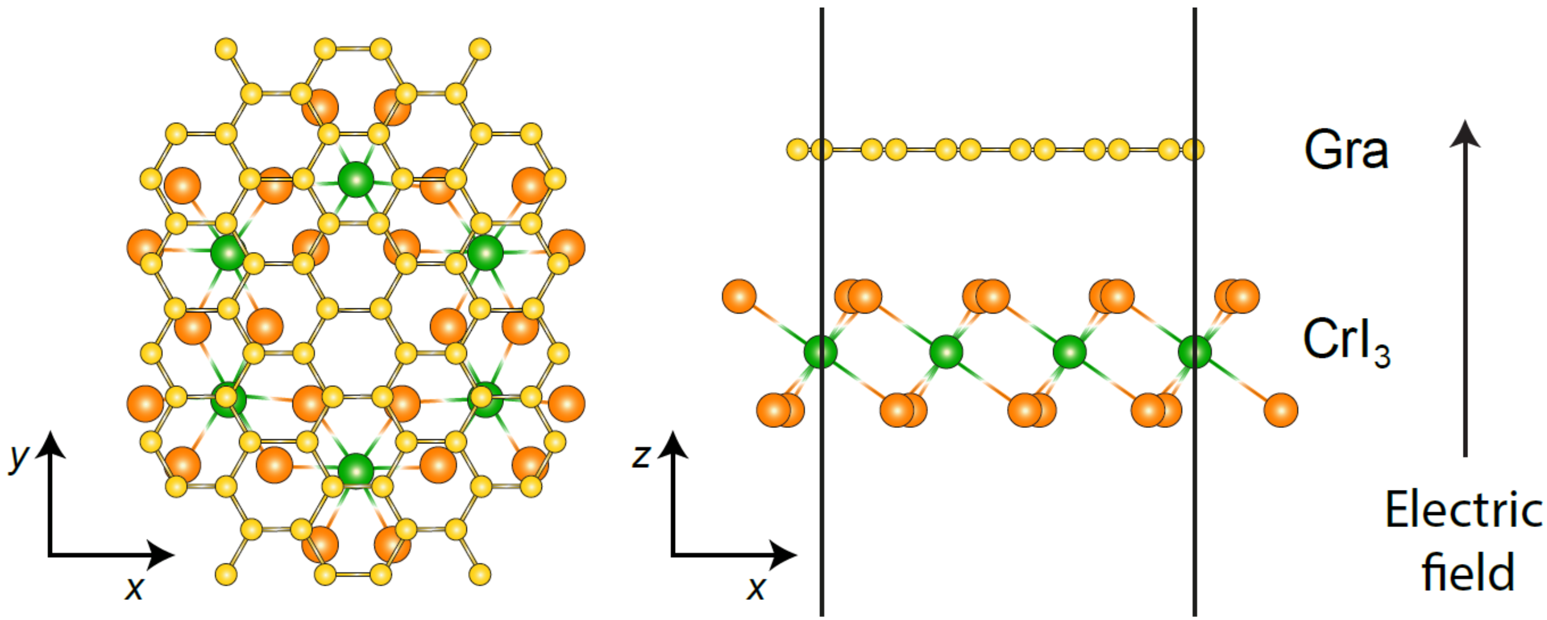
Calculated MAE



- The MAE increases (decreases) with Fe (Sc) concentration.
- the MAE can be effectively adjusted by Fe or Sc doping without compromising the insulating property of CrI_3.

Proximity to graphene

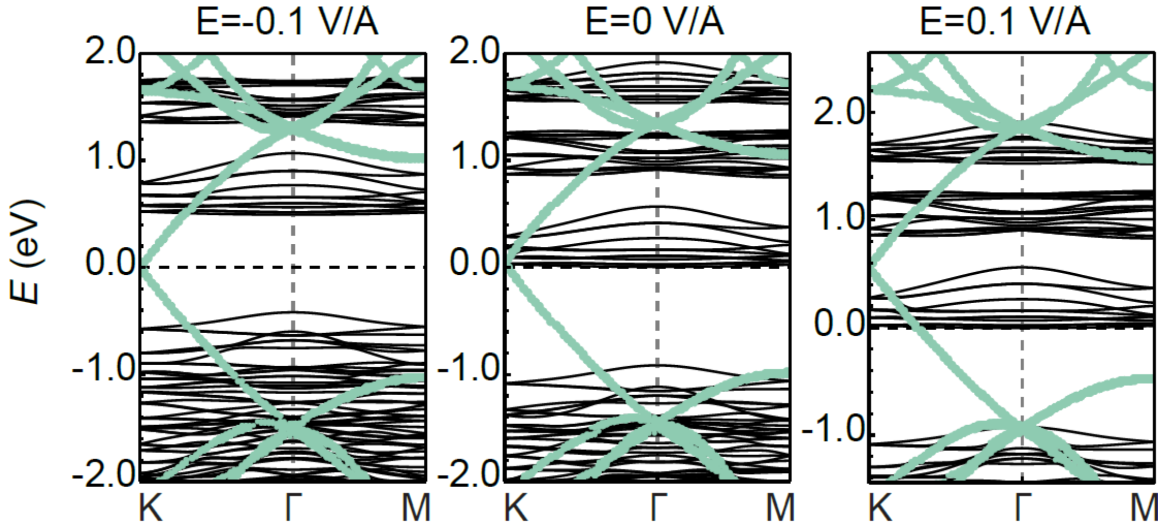
Geometry



a (5×5) graphene is placed on a (√3×√3) CrI_3 monolayer

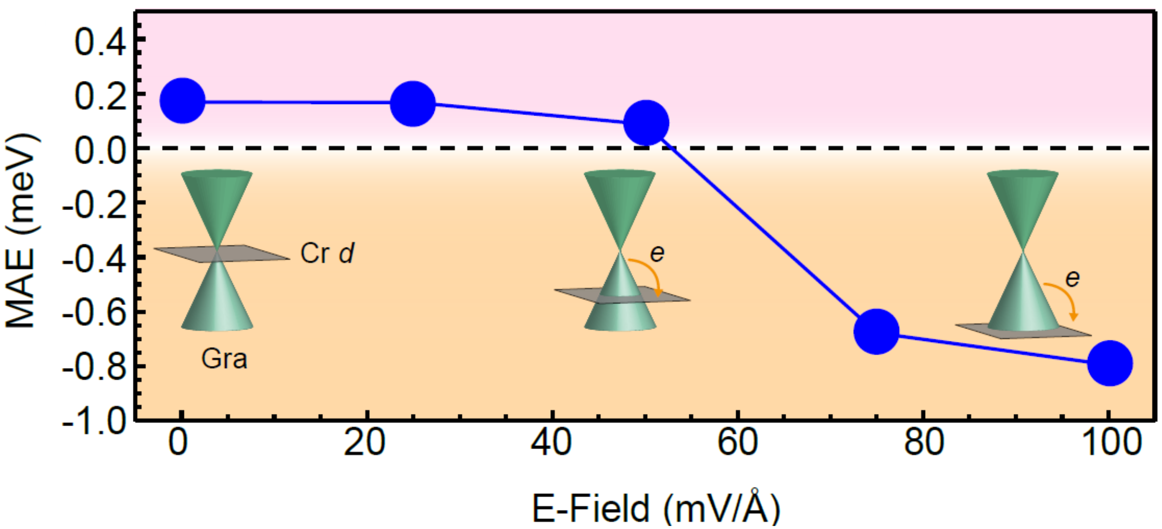
Proximity to graphene

Band structure



- Charge transfer is driven by the perpendicular external field.

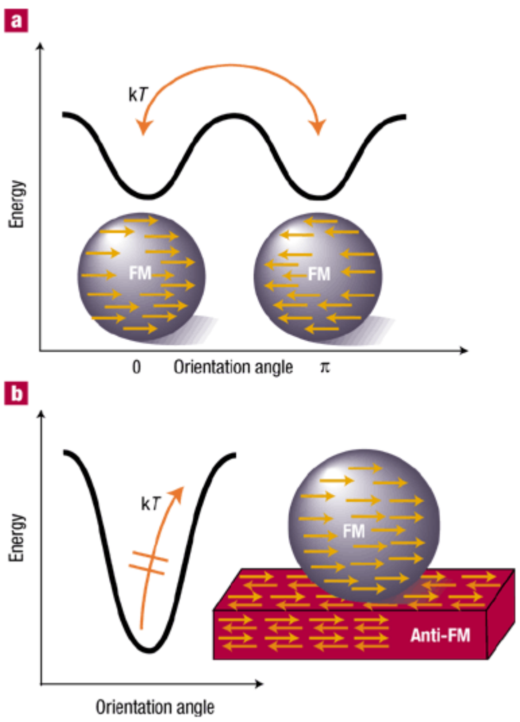
Calculated MAE



- The spin configuration of CrI_3 can be dynamically controlled by using a perpendicular external field

Proximity to graphene

Dynamical manipulation of MAE



For switching, the energy barrier can be instantaneously vanished by electric field as the magnetic states is reconfigured by nearly zero current.

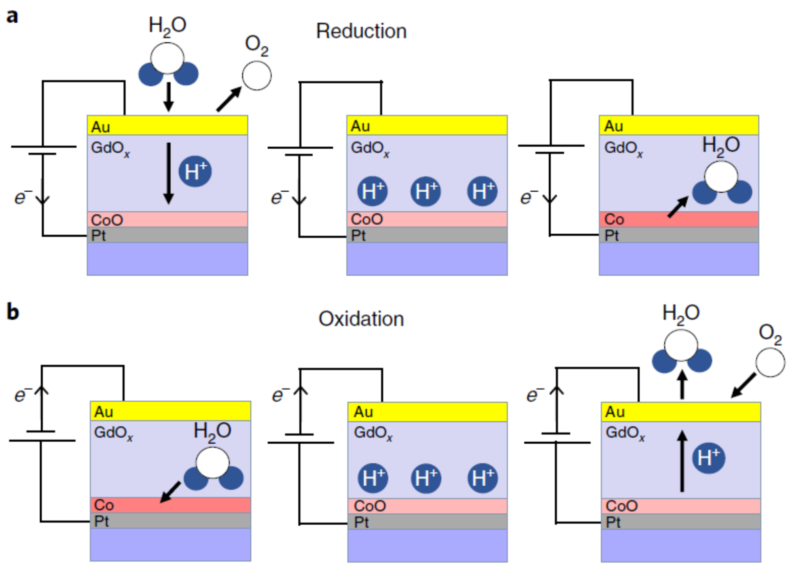
For stability, the energy barrier is recovered by removing electric field to retain a magnetic state.

J Eisenmenger *et al*., *Nat. Mater.* 2 437 (2003)

⇨ **energy-efficient spin device**

Previous magnetic systems

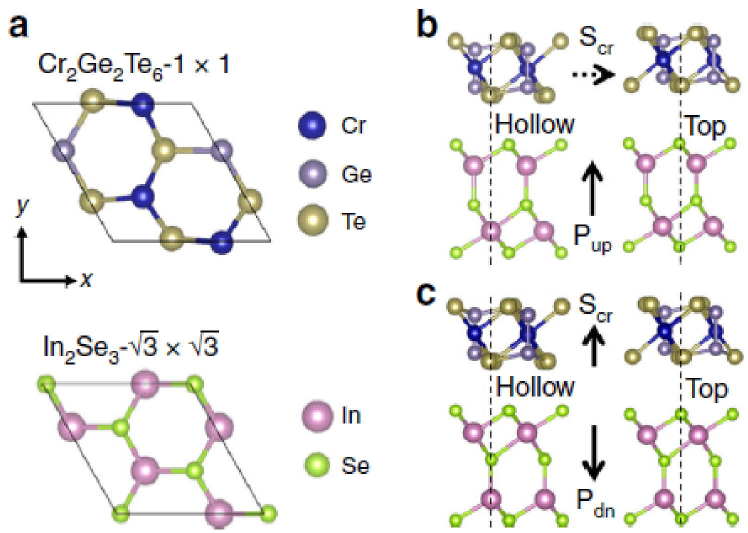
Magneto-ionic control



A. J. Tan *et al.*, *Nat. Mater.* 18 35 (2019)

- Large variation of MAE, but a long switching time form microseconds to minutes.
- Our charge-transfer mechanism is very fast.

Multiferroicity



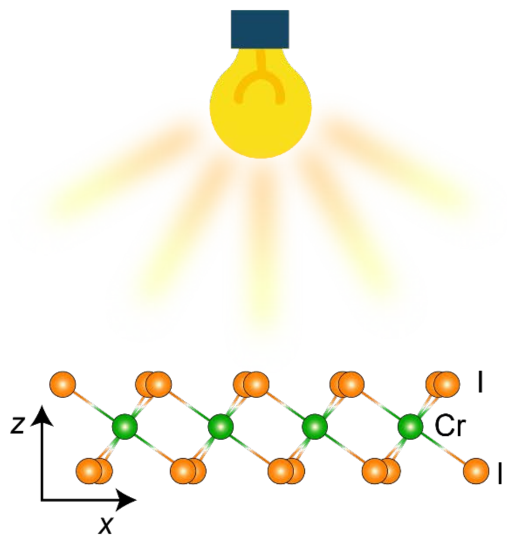
C. Gong *et al.*, *Nat. Commun.* 10 1 (2019)

- Difficult to tune the magnitude of MAE
- A wide range of MAE can be accessible by adjusting electric field in our case.

High-speed and reversible spin device

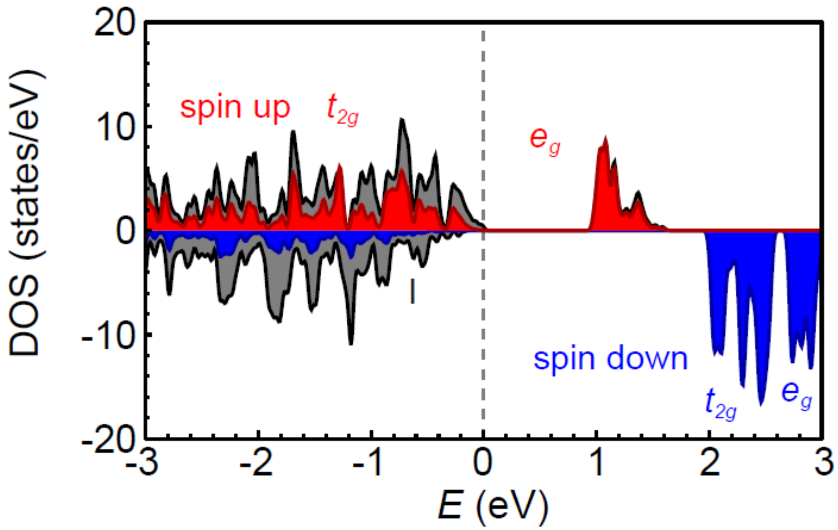
Optical excitation

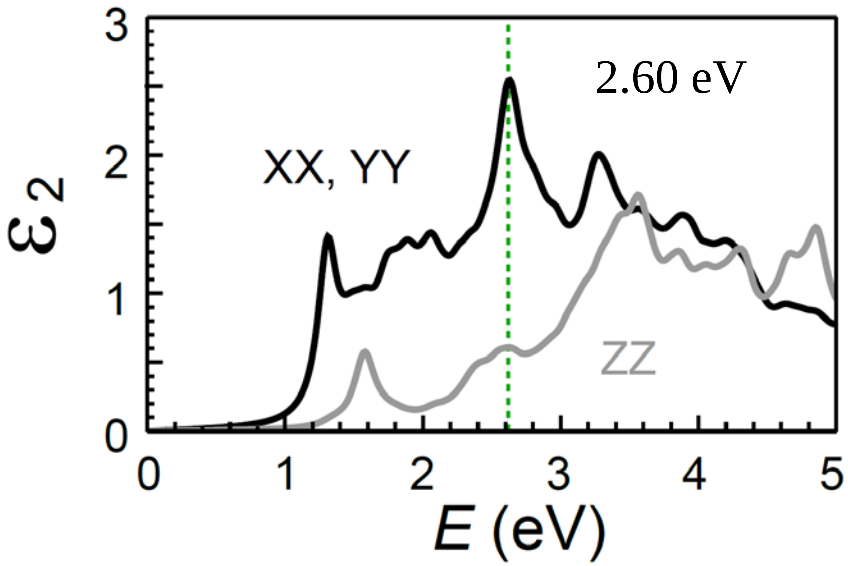
Carrier excitation through irradiation



We consider the manipulation of the MAE of **a pristine and stand-alone CrI_3** monolayer through electronic excitation obtained by using a resonant laser pulse.

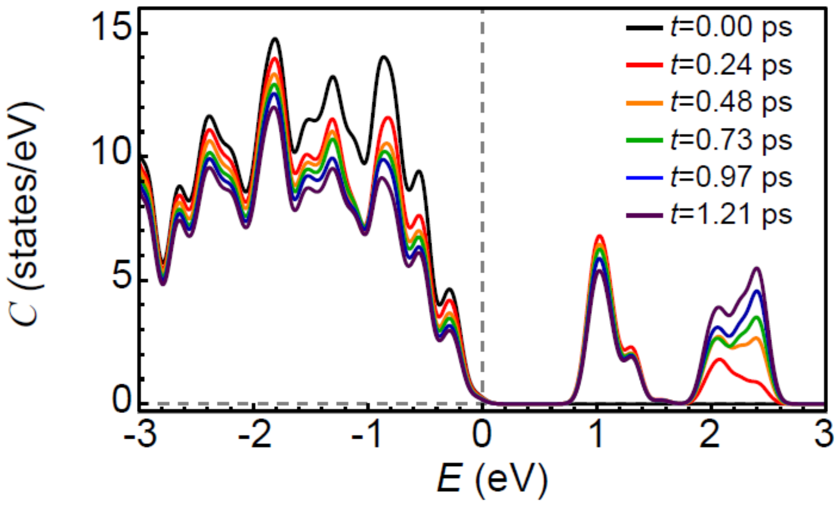
Choice of Light frequency





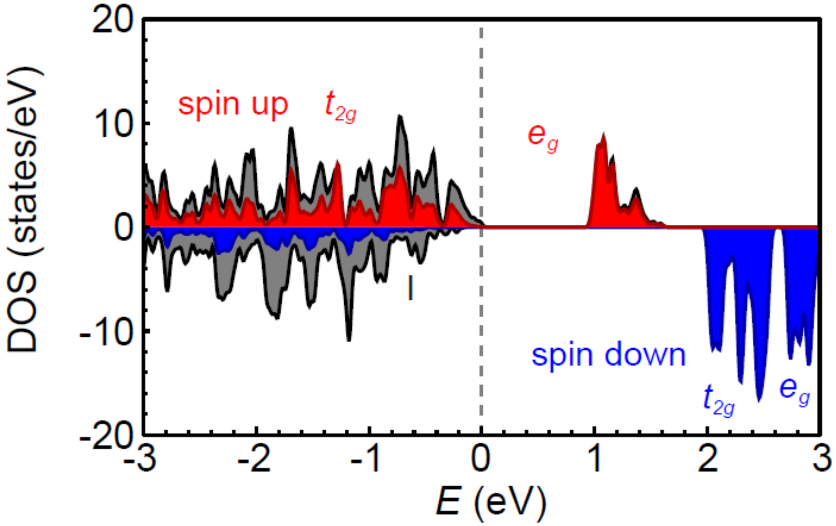
Optical excitation

Time-varying DOS of occupied states



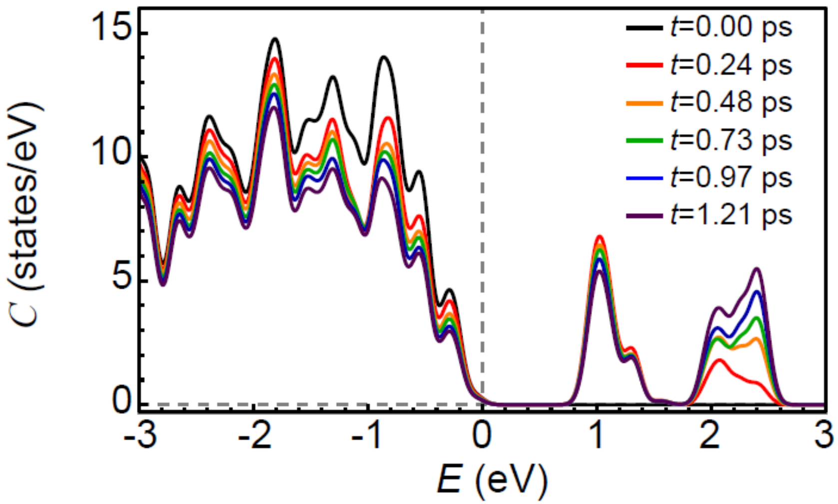
$(C(E,t) = d|\langle\psi(t)\rangle|\psi(0)\rangle|^2/dE)$

t=0



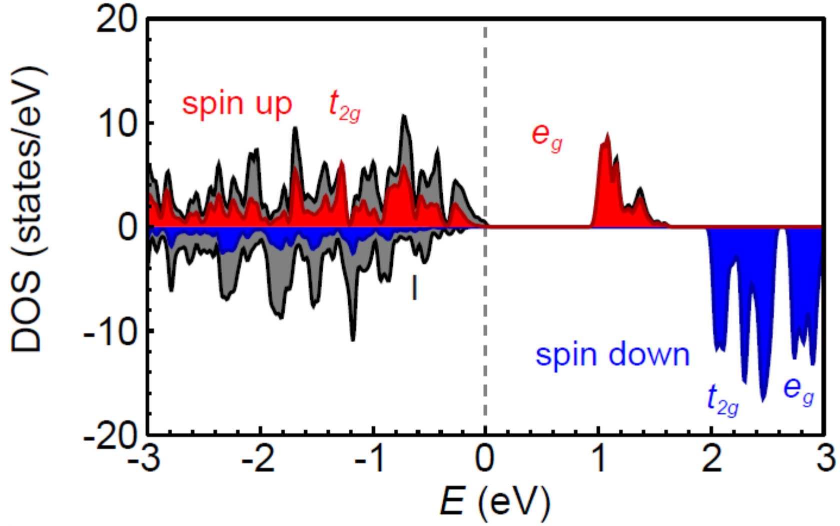
Optical excitation

Time-varying DOS of occupied states

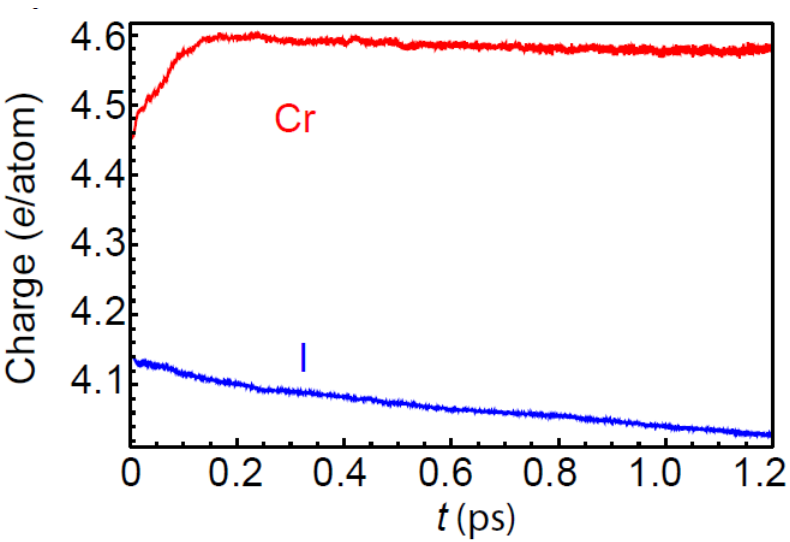


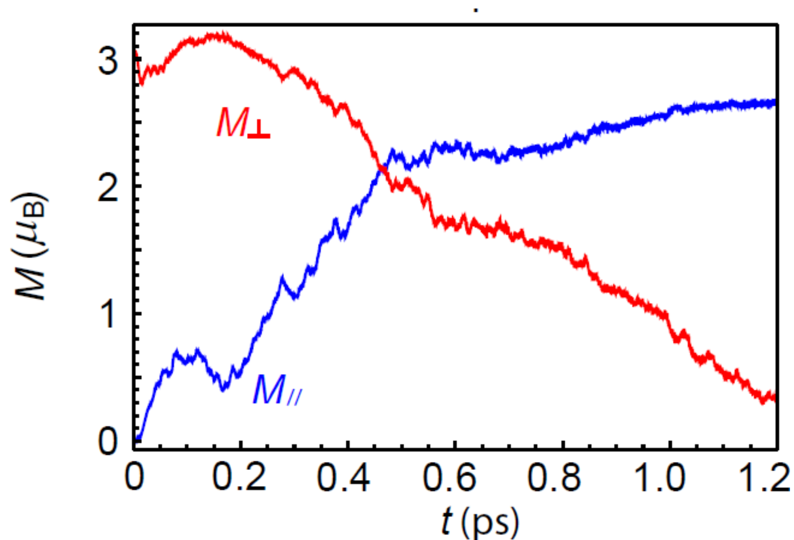
$(C(E,t) = d|\langle\psi(t)\rangle|\psi(0)\rangle|^2/dE)$

***t*=0**



Reorientation of spins





Summary

1. We performed first-principles calculations for MAE in CrI_3-based 2D structures and presented a few accessible methods to manipulate its magnetization configurations.

2. As a result of extensive DFT calculation, we found that, in the determination of the magnetization configuration, the spin and orbital angular momentums between the occupied and unoccupied bands are intertwined, giving rise to the drastic variation of the MAE upon the band filling

3. Especially, we demonstrated that the anisotropic magnetic configuration can be easily altered by the perpendicular external field, implying high potential for energy-efficient fast switching of two-dimensional magnets with nearly zero critical current density.